A Viscosity Test Method using a Search Coil Detection System and Superparamagnetic Nanoparticles

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ABSTRACT

We propose and demonstrate a Brownian relaxation based mixing frequency method to test sample viscosities. This method uses excitation and detection coils and Brownian relaxation dominated superparamagnetic nanoparticles (MNPs), which are sensitive to the liquid environment such as viscosity, hydrodynamic volume, and temperature. A low frequency sinusoidal magnetic field is applied to saturate the MNPs into nonlinear region and a high frequency field is applied to modulate it into the high frequency region where the noise floor is lower. The phase lag of the 3rd harmonic and induced voltage ratio of the 5th over the 3rd harmonic signals are collected. We build up standard graphs by putting collected data from eight MNP mixtures with different viscosities. For any unknown liquids mixed by MNPs, we can collect phase lag and voltage ratio information and insert these data into aforementioned standard graphs. This in vitro viscosity test can be done in 1 minute. Our experimental result showed a 0.3% error rate.

I. BACKGROUND

There exist two relaxation mechanisms for MNPs under alternating magnetic fields: Brownian and Néel relaxation. Brownian relaxation is the physical rotation of MNPs in solution and Néel relaxation is the magnetic dipole flipping inside the particle. Brownian and Néel relaxation times are expressed as:

$$\tau_B = \frac{3\eta V_H}{k_B T} \qquad \tau_N = \tau_0 \exp\left(\frac{KV_m}{k_B T}\right)$$

Respectively, where η is the viscosity of the MNP solution, V_H is the hydrodynamic volume, k_b is the Boltzmann constant; and T is the temperature of the solution in Kelvin. $\tau_0 = 10^{-9}$ s, K_u is the uniaxial energy density, and V_m is the magnetic core volume of each MNP. Since SHP-25 has a core diameter of 25nm, the dominant relaxation time would be Brownian. Using this information, the viscosity of any unknown liquid can be determined, as long as we collect voltage ratio and phase lag information.

The search coil system uses three coils: one to for a high frequency magnetic field, one for a low frequency magnetic field, and a pair of differentially rounded detection coils. The magnetic field can be described as:

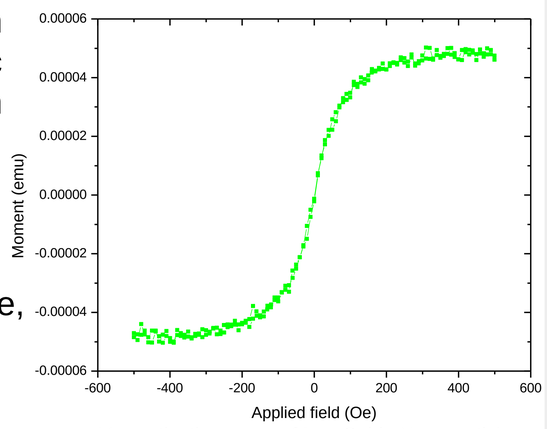
$$H = A_L \cos(2\pi f_L t) + A_H \cos(2\pi f_H t)$$

Phase lag ϕ can be expressed as:

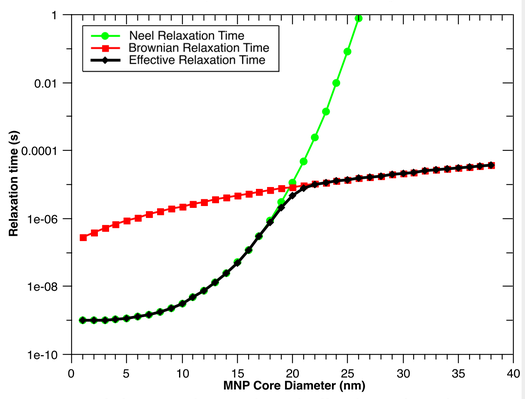
$$\phi = \arctan(\omega\tau)$$

The voltage ratio of the 5th harmonic over 3rd harmonic can be expressed using Lenz law:

$$\frac{V_{5th}}{V_{3rd}} \propto \frac{\omega_{5th}}{\omega_{3rd}} \cdot M_0^2 \cdot \frac{\cos\phi_{5th}}{\cos\phi_{3rd}} \propto \left(M \cdot \cos\phi\right)^2$$



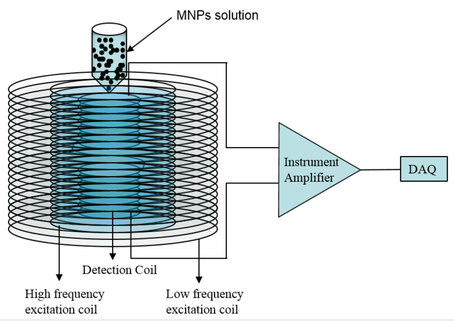
Magnetization curve of Iron Oxide Nanoparticles (SHP-25, Ocean NanoTech) in water solution at room temperature measured by VSM.



Néel, Brownian and total effective relaxation time as function of MNP core size, T=293K, viscosity of MNP solution is 1 cp.

II. OBJECTIVE

- Collect amplitudes and phases of 3rd and 5th harmonics from the detection coils.
- Viscosity of unknown liquids were determined using voltage ratios and phases



III. EXPERIMENTAL METHODS

- Digital acquisition card (DAQ, NI USB-6289, 18-Bit, 625 kS/s), LabVIEW and Matlab are used for instrument control and signal processing, respectively.
- 50μL SHP-25 nanoparticles (purchased from Ocean NanoTech, iron oxide core MNPs of 25nm diameter) were mixed with 200μL varying glycerol and distilled water combinations.
- Their voltage ratio and phase lag data was collected by inserting the MNP solutions into the center of the coil. Three trials of varying high frequency fields (300 Hz, 500 Hz, 900 Hz) were used. The theoretical viscosity of the mixtures were determined using this data.
- Actual viscosities of the mixtures were determined using an AR-G2 rheometer

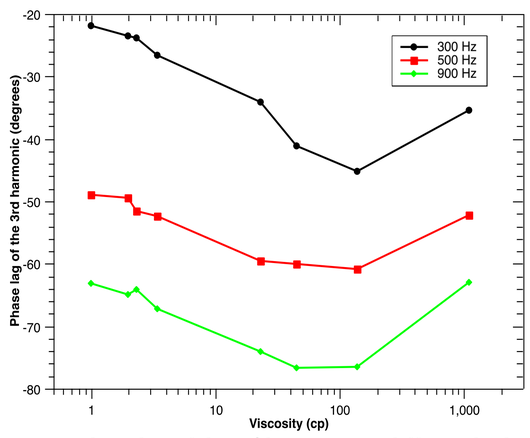
IV. RESULTS AND DISCUSSION

The eight mixtures of 200 μL of varying glycerol concentrations were prepared and the viscosities were found using an AR-G2 rheometer. These are the table of standard values:

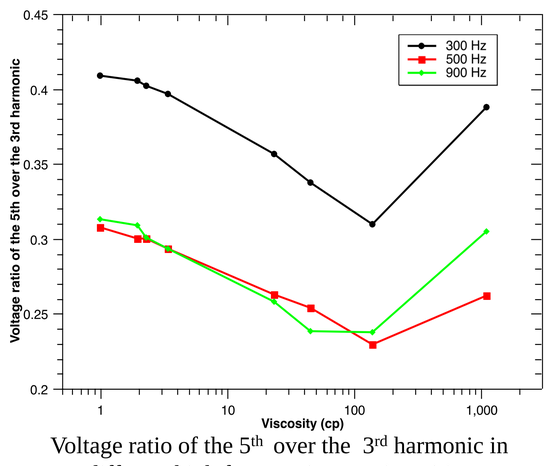
Mixture	Viscosity	Mixture	Viscosity
A	0.99 cp	E	22.99 cp
B	1.95 cp	F	44.78 cp
C	2.29 cp	G	136.58 cp
D	3.36 cp	H	1087.43 cp

Viscosities of 8 samples at 20°C

The phase and voltage ratio signals collected from these samples. Standard graphs were built:



Phase lag of the 3rd harmonic in different high frequencies vs. viscosities



Voltage ratio of the 5th over the 3rd harmonic in different high frequencies vs. viscosities

Two mixtures of unknown viscosities were tested. Mixture I contained 200μL glycerol and DI water solution mixed with 50μL MNP, and mixture II contained 200μL serum (male human serum type AB, purchased from Sigma-Aldrich Company, this product consists of hemoglobin ≤ 20mg/dL, and endotoxin ≤ 10EU/mL) mixed with 50μL MNP. The phase lag and voltage ratio data was collected and compared with the standard graph. The viscosity was determined using the graph.

	Methods	Phase lag of the 3rd harmonic			Voltage ratio		
	Frequency	300Hz	500Hz	900Hz	300Hz	500Hz	900Hz
Mixture I	Raw Data	-30.52	-54.34	-71.18	0.384	0.278	0.278
	Viscosity	9.0	7.0	8.0	7.6	8.4	7.8
Mixture II	Raw Data	-38.42	-52.72	-68.89	0.394	0.384	0.376
	Viscosity	35	2.8	5.2	2.8	NA	NA

Phase lag of the 3rd harmonic, voltage ratio of the 5th over the 3rd harmonics, tested at 20°C

The tested viscosity of Mixture I is estimated to be 7.97 cp, and compared to the actual viscosity the error rate turned out to be 0.3%. However, an abnormal phenomenon occurred with mixture II. The estimated viscosity is at least 60% higher than the actual value. This may be because the proteins in the human serum were nonspecifically conjugated on the surfaced of the MNPs and therefor increased the hydrodynamic volume of the MNPs. This in turn would increase the effective relaxation time and make the viscosity appear that it is higher than the theoretical viscosity value.

V. CONCLUSIONS

- A search coil detection system with superparagmagnetic nanoparticles was used to estimate viscosities of unknown solutions
- There was a 0.3% error rate when estimating unknown viscosities of glycerol and distilled water and human serum and blood viscosity, given that the MNPs do not conjugate (using a PEG outer layer covered MNPs)
- Compared to other viscosity testing methods (like micro electro mechanical system), our search coil system far more cheaper and efficient.

ACKNOWLEDGMENTS

1. Q. A. Pankhurst, J. Connolly, S. Jones and J. Dobson, Journal of physics D: Applied physics **36** (13), R167 (2003).
2. O. Cakmak, C. Elbuken, E. Ermek, A. Mostafazadeh, I. Baris, B. Erdem Alaca, I. H. Kavakli and H. Urey, Methods **63** (3), 225-232 (2013).
3. O. Cakmak, E. Ermek, H. Urey, G. G. Yaralioglu and N. Kilinc, presented at the Sensors, 2013 IEEE, 2013 (unpublished).
4. A. M. Rauwerdink and J. B. Weaver, Applied Physics Letters **96** (3), 033702 (2010).
5. S.-H. Chung, A. Hoffmann, K. Guslienko, S. Bader, C. Liu, B. Kay, L. Makowski and L. Chen, Journal of applied physics **97** (10), 10R101 (2005).
6. M. Lucchini and F. Canepa, Journal of Nanoparticle Research **14** (4), 1-7 (2012).
7. L. Tu, K. Wu, T. Klein and J.-P. Wang, Journal of Physics D: Applied Physics **47** (15), 155001 (2014).
8. K. Wu, Y. Wang, Y. Feng, L. Yu and J.-P. Wang, Journal of Magnetism and Magnetic Materials (2014).
9. K. Wu, L. Yu, X. Zheng, Y. Wang, Y. Feng, L. Tu and J.-P. Wang, presented at the Engineering in Medicine and Biology Society (EMBC), 2014 36th Annual International Conference of the IEEE, 2014 (unpublished).
10. A. M. Rauwerdink, E. W. Hansen and J. B. Weaver, Physics in medicine and biology **54** (19), L51 (2009).
11. J. B. Weaver, A. M. Rauwerdink and E. W. Hansen, Medical physics **36** (5), 1822-1829 (2009).
12. J. B. Weaver, M. Harding, A. M. Rauwerdink and E. W. Hansen, presented at the SPIE Medical Imaging, 2010 (unpublished).
13. L. Tu, Y. Feng, T. Klein, W. Wang and J.-P. Wang, Magnetics, IEEE Transactions on **48** (11), 3513-3516 (2012).
14. L. Tu, T. Klein, W. Wang, Y. Feng, Y. Wang and J.-P. Wang, Magnetics, IEEE Transactions on **49** (1), 227-230 (2013).
15. H.-J. Krause, N. Wolters, Y. Zhang, A. Offenhäusser, P. Miethe, M. H. Meyer, M. Hartmann and M. Keusgen, Journal of Magnetism and Magnetic Materials **311** (1), 436-444 (2007).
16. P. I. Nikitin, P. M. Vetoshko and T. I. Ksenevich, Journal of Magnetism and Magnetic Materials **311** (1), 445-449 (2007).
17. J. B. Weaver and E. Kuehlert, Medical physics **39** (5), 2765-2770 (2012).
18. J. Dieckhoff, M. Schilling and F. Ludwig, Applied Physics Letters **99** (11), 112501 (2011).